Spin Wave Response in the Dilute Quasi-one Dimensional Ising-like Antiferromagnet $CsCo_{0.83}Mg_{0.17}Br_3$

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Abstract

Inelastic neutron scattering profiles of spin waves in the dilute quasi-one-dimensional Ising-like antiferromagnet $CsCo_{0.83}Mg_{0.17}Br_3$ have been investigated. Calculations of $S^{xx}(Q,\omega)$, based on an effective spin Hamiltonian,

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accurately describe the experimental spin wave spectrum of the $2J$ mode. The Q dependence of the energy of this spin wave mode follows the analytical prediction $\omega_{xx}(Q)$=(2J)(1-5$\epsilon^2 cos^2 Qa + 2\epsilon^2)^{1/2}$, calculated by Ishimura and Shiba using perturbation theory.

The quantum nature of spin-1/2 one-dimensional magnetic systems manifests itself in a number of interesting properties that have been studied both experimentally and theoretically for many years [1]. The spin wave excitation spectrum around the pure Ising energy consists of a continuum of states, accompanied by the propagation of domain wall pairs of the soliton response. The existence of a band of spin wave states and the soliton response have been confirmed within the quasi-one-dimensional system, $CsCoM_3$(M=Br, Cl), by Satija *et al.* [2] and Nagler *et al.* [3].

The spin Hamiltonian describing the dynamics of 1-D Ising-like antiferromagnets, including the exchange mixing produced by inter-chain and intra-chain interactions, was determined to be [4]

$$H = 2J \sum [S_j^z S_{j+1}^z + \epsilon (S_j^x S_{j+1}^x + S_j^y S_{j+1}^y)] + h \sum (-1)^j S_i^z. \qquad (1)$$

Here J is the exchange coupling between nearest neighbour Co ions in a chain, ϵ is the parameter that distinguishes the Ising limit ($\epsilon = 0$) from the Heisenberg limit ($\epsilon = 1$), h is an effective (staggered) field, which arises from exchange mixing and coupling between chains.

The Hamiltonian (1) may also model the case where a non-magnetic ion (Mg) is substituted for the magnetic ion (Co), which gives a distribution of finite length spin chains. Finite length chains are of interest because they allow one to study surface modes with neutron scattering. Moreover, comparison with theory is more rigorous since the full spectrum, $S^{xx}(Q, \omega)$, can be computed exactly for Hamiltonian (1) in small chains. Nagler *et al.* [5] measured the spin wave spectrum of such a dilute one-dimensional chain, $CsCo_{0.83}Mg_{0.17}Cl_3$, with inelastic neutron scattering and carried out a detailed comparison with theoretical results based on such a Hamiltonian. Their results confirmed the necessity of the staggered

field. Nonetheless, the calculated spin wave spectrum at the zone center was in disagreement with experiment, and the relative intensities between the surface mode ($\omega \sim J$) and the bulk mode ($\omega \sim 2J$) were not in quantitative agreement. Furthermore, only two wave vectors were examined, therefore up until the present, a full experimental study of the spin wave dispersion has been lacking. Such a dispersion relation was calculated by Ishimura and Shiba (IS) [7], and it remains an unchallenged prediction relevant to the effective Hamiltonian described above.

In this paper, by measuring the spin wave response for several Q-vectors throughout the Brillouin zone, it is shown that a refined calculation of $S^{xx}(Q,\omega)$ quantitatively describes the spin wave spectrum associated with both the J and the $2J$ mode, and that the Q-dependence of the spin wave energy of this mode indeed follows the IS prediction:

$$\omega_{xx}(Q) = (2J)(1 - 5\epsilon^2 cos^2 Qa + 2\epsilon^2)^{1/2}. \tag{2}$$

$CsCoBr_3$ belongs to the space group D^4_{6h} and displays room temperature lattice constants a=7.529 $\mathring{A}$ and c=6.324 $\mathring{A}$. The material consists of stacked triangular lattices composed of chains of Co^{++} ions arranged parallel to the c-axis. The Co^{++} ions on a given chain interact strongly with one another, with exchange constant J. Long range, three-dimensional ordering occurs at $T_N = 28.4$ K, below which $CsCoBr_3$ enters a partially-paramagnetic, 3-sublattice Neel state. On further cooling, an additional transition occurs, associated with the ordering of the paramagnetic site. With the addition of non-magnetic impurities, Mg (17%), in place of Co, the ordered states are severely altered in such a way that order parameter measurements show upwards curvature to temperatures as low as 3 K [8]. In our measurements we stay well above these temperatures, with the intent of avoiding three dimensional and ordering effects.

$CsCo_{1-x}Mg_xBr_3$ is a good candidate system for the investigation of dilution effects as the properties of the pure system have been well determined. In addition, for neutron studies, the bromide is expected to be preferable to the chloride as the incoherent scattering cross section of Br is considerably less than Cl (0.1 barns compared with 5.1). In addition the

absorption cross-section is considerably lower for Br than for Cl. The dilute system allows for study of both the $2J$ mode, which involves a flip of a spin in the bulk of a finite chain as well as for the J mode which arises from spin flips that occur at either end of the finite chains.

Inelastic neutron scattering experiments were carried out on the HB2 triple axis spectrometer at the HFIR research reactor of ORNL. Measurements were made with fixed final neutron energy 3.52 THz, using Si(111) as the monochromator and pyrolytic (002) as the analyzer. Collimation of 40' was chosen for both the incident and scattered beams. The resulting energy resolution was about 0.25 THz. A pyrolytic graphite filter was used in the scattered beam to suppress the higher order contamination.

For the dilute system, the transverse spin wave response $S^{xx}(Q,\omega)$can be written in terms of the response function for a single chain of length N, $S_N^{xx}(Q,\omega)$:

$$S^{xx}(Q,\omega) = \sum_N u^N (1-u)^2 S_N^{xx}(Q,\omega), \tag{3}$$

where the single chain response is

$$S_N^{xx}(Q,\omega) = \sum_E |< E|S_N^{xx}(Q)|G>|^2 \delta(\omega - E). \tag{4}$$

Here the spin-flip operator is given by

$$S_N^{xx}(Q,\omega) = \sum_{j=1}^{N} e^{iQj} \mathbf{S}_j^x, \tag{5}$$

where the summation of j is over the length of the chain, and $\mathbf{S}_j^x$ is the usual x-component of the spin operator for the spin at the jth site. In these expressions, we have used u to denote the concentration of magnetic ions. The summation over the various chain lengths is therefore subject to the constraint that the total number of magnetic ions in the entire chain equals this concentration. We use the ket $|E>$ to denote an eigenstate with energy E and $|G>$ is the ground state with energy taken to be zero.

The distribution of chain lengths is assumed to be based on random substitution of non-magnetic Mg ions for Co in the chain. This is the same percolation assumption used by Nagler *et al.* [5] for $CsCo_{0.83}Mg_{0.17}Cl_3$.

The total $S^{xx}(Q,\omega)$ for the system is then obtained by first adding the response of chains with $N \leq 11$, weighted according to the above-mentioned percolation theory. The resulting response is then scaled from $N = 11$ to larger sizes by multiplying the computed response by two factors, $F_{2J}^{\infty}/F_{2J}^{11}$ for the $2J$ band and $F_{J}^{\infty}/F_{J}^{11}$ for the J band. The scaling functions are

$$F_{2J}^{M} = \sum_{N=2}^{M} (N-2)(1-u)^2 u^N, \;\; F_{J}^{M} = \sum_{N=2}^{M} 2(1-u)^2 u^N, \tag{6}$$

and are given by [5]

$$F_{2J}^{M} = x^3 - x^{M+1}[(M-2)(1-x)+1] \tag{7a}$$

and

$$F_{J}^{M} = 2x^2(1-x)(1-x^{M-1}). \tag{7b}$$

The F-functions with $M = \infty$ are those that would be obtained from an Ising system (at $T = 0$), with an average number of chains of length N equal to $x^N(1-x)^2$ (according to percolation theory [5]). The calculated responses up to some M ($M = 11$ in our case) are weighted by these factors in order to account for the presence of larger chains (which is non-negligible).

The spin wave response at 13.3 K in $CsCo_{0.83}Mg_{0.17}Br_3$, is shown in Fig. 1, for a variety of wave vectors spanning the one-dimensional Brillouin zone from the zone boundary (0, 0, 2.5) to the zone centre (0, 0, 3). All the constant **Q** scans taken were of the form (0, 0, L) and sensitive only to transverse spin correlations, hence $S^{xx}(Q,\omega)$, due to the sensitivity of the neutron scattering cross section to spin components normal to **Q**. The experimental and calculated peak intensities have been scaled to agree at a single wavevector, (0, 0, 2.5). The data shows two clearly defined modes with energy J and $2J$, which are identified as magnetic in origin, both by their fall off at large wave vector due to the magnetic form factor and by the temperature dependence of this scattering. The phonon peaks, which were also found for the pure sample [4], are indicated by the letter P.

The solid curves are calculations of $S^{xx}(Q,\omega)$ weighted, as described above, for chains with $N \leq 11$. We checked the appropriate scaling relations by calculating the spectrum

for $N \leq 14$, and utilizing the same procedure. The results changed very little; indeed, the maximum spin chain length originally utilized by Nagler *et al.* [5] ($N \leq 8$) also gave a fairly accurate representation of the full distribution. This result is not surprising as the average chain length is 5. These spectra were calculated at $T = 13.3$ K, with parameter values similar to those used to describe the pure sample, $J = 1.55THz$, and $\epsilon = 0.18J$ [3]. The staggered field parameter $h = 0.05J$ and a convolution of the resolution function of the instrument with the expected Lorentzian function [6] with full width at half maximum $\Delta = 0.1J$ for the spin wave peak were used without further adjustable parameters.

Typically the value of the exchange constant J is determined by the peak position of the $2J$ mode at the zone boundary. However, as the peak positions of the two modes depend on both h and ϵ as well, we have fit the spectrum for J, h and ϵ simultaneously to give optimal agreement at all wave vectors studied. A slight discontinuity occurs in the calculated curves in Fig. 1 at an energy between the J and $2J$ modes ($1.5J$). This artifact arises because of the different scaling employed for each frequency range, as discussed above. As can be seen from Fig. 1 the description of the experiment by theory is quantitatively very good at all wavevectors across the 1D Brillouin zone.

IS calculated the dispersion for the $2J$ mode, using a combination of perturbation theory and the previously known second moment result [7], for the transverse spin wave response in the absense of a staggered field term. Their resulting prediction is Eq. (2). We have evaluated the energies corresponding to the center of mass of the calculated spectra for all wave vectors in Fig. 1 and have found that the ω-Q dispersion is indeed well described by IS theory. In the figure, the energies corresponding to the center of mass are slightly larger than the absolute values calculated from Eq. (2). However, this is largely due to the influence of the staggered field which raises the energy of the spectra by about $0.13J$ compared with the h=0 case at the zone boundary. It is worth emphasizing that the excellent description of the scattering line shape by the calculated structure factor implies the IS energy dispersion for the $2J$ mode. These results clearly indicate that the effective Hamiltonian (1) describes very accurately the dispersion of the $2J$ mode.

The dispersion relation obtained from the fits in Fig. 1 is shown in Fig. 2. The open circles ($2J$ mode) are the energies of the center of mass of the spectra. The filled circles show the $2J$ mode energies corrected for the effect of finite staggered field so that they can be compared directly with Eq. (2). The solid line represents the IS theoretical expression, Eq. (2). In this figure, the squares show the dispersion of J mode using the same procedure as was used for $2J$ mode energies.

In Fig. 3, we show a two dimensional contour map of the calculated inelastic spectrum as a function of energy and wave vector. In this figure, the parameters used are the same as those employed in the fits to the data shown in Fig. 1. This scattering shows the spin wave continuum as predicted by IS, giving the antisymmetric bow-tie shape for the $2J$ mode. The spin wave continuum broadens appreciably as the zone center is approached, to the values $\sim \pm 4\epsilon J$, consistent with previous theortical work [7,9].

Fig. 4 shows a two dimensional contour map of the calculated spin wave spectrum as a function of staggered field, for the zone center wavevector. The pronounced increase in the ratio of intensities between the $2J$ and J modes, as well as the movement of spectra towards higher energy with increasing h can be clearly seen. Both the description of the ratio of the relative intensities in the $2J$ and J modes in the experimental spin wave scattering intensity, as well as the description of the asymmetry of the spin wave lineshapes have been improved by adjusting h and ϵ.

There has been some debate regarding the use of a next nearest neighbour exchange interaction (NNN) for the 1-D system in the pure sample [10,11]. We have investigated this effect and found the NNN term is not required [11] for a satisfactory description of the data. We have seen that the staggered field plays a key role in obtaining the very good correspondence between the experimental and the calculated lineshapes.

To conclude, by measuring the spin wave response for a variety of scattering wavevectors, we have shown that a quantitative understanding of the transverse spin wave spectrum $S^{xx}(Q,\omega)$ in the quasi-one-dimensional, diluted spin-1/2 chains system $CsCo_{0.83}Mg_{0.17}Br_3$ can be obtained through an effective spin Hamiltonian (1). This result provides a stringent

test of the IS perturbative form for the dispersion relation; by attempting fits without the staggered field term (and failing), we have highlighted the importance of such a term, as first suggested by Nagler *et al.* [4].

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REFERENCES

[1] D.C. Mattis, *The Many-Body Problem*, (World Scientific, Singapore, 1993).

[2] S. K. Satija, G. Shirane, Y. Yoshizawa and K. Hirakawa, Phys Rev. Lett. **44** 1548 (1980).

[3] S. E. Nagler, W. J. L. Buyers, R. L. Armstrong and B. Briat, Phys. Rev. Lett. **49** 590 (1982).

[4] S. E. Nagler, W. J. L. Buyers, R. L. Armstrong and B. Briat, Phys. Rev. B**27** 1784 (1983).

[5] S. E. Nagler, W. J. L. Buyers, R. L. Armstrong and R. A. Ritchie, J. Phys. C:Solid State Phys. **17** 4819 (1984).

[6] J. Villain(p91), M. Steiner(p107), *Ordering in Strongly Fluctuating Condensed Matter System*, edited by T. Riste (Plenum, New York, 1980).

[7] N. Ishimura and H. Shiba, Prog. Theoretical Phys. **63** 743 (1980).

[8] R. B. Rogge, Y. S. Yang, Z. Tun, B. D. Gaulin, J. A. Fernandez-Baca, R. M. Nicklow and A. Harrison, J. Appl. Phys. **73**6451 (1993).

[9] J. des Cloizeaux and M. Gaudin, J. Math. Phys. 7, 1384 (1966)

[10] F. Matsubara and S. Inawashiro, Phys. Rev. B **43** 1, 796 (1991).

[11] J. P. Goff, D. A. Tennant and S. E. Nagler, Phys. Rev. B **52** 22, 15992 (1995).

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FIGURES

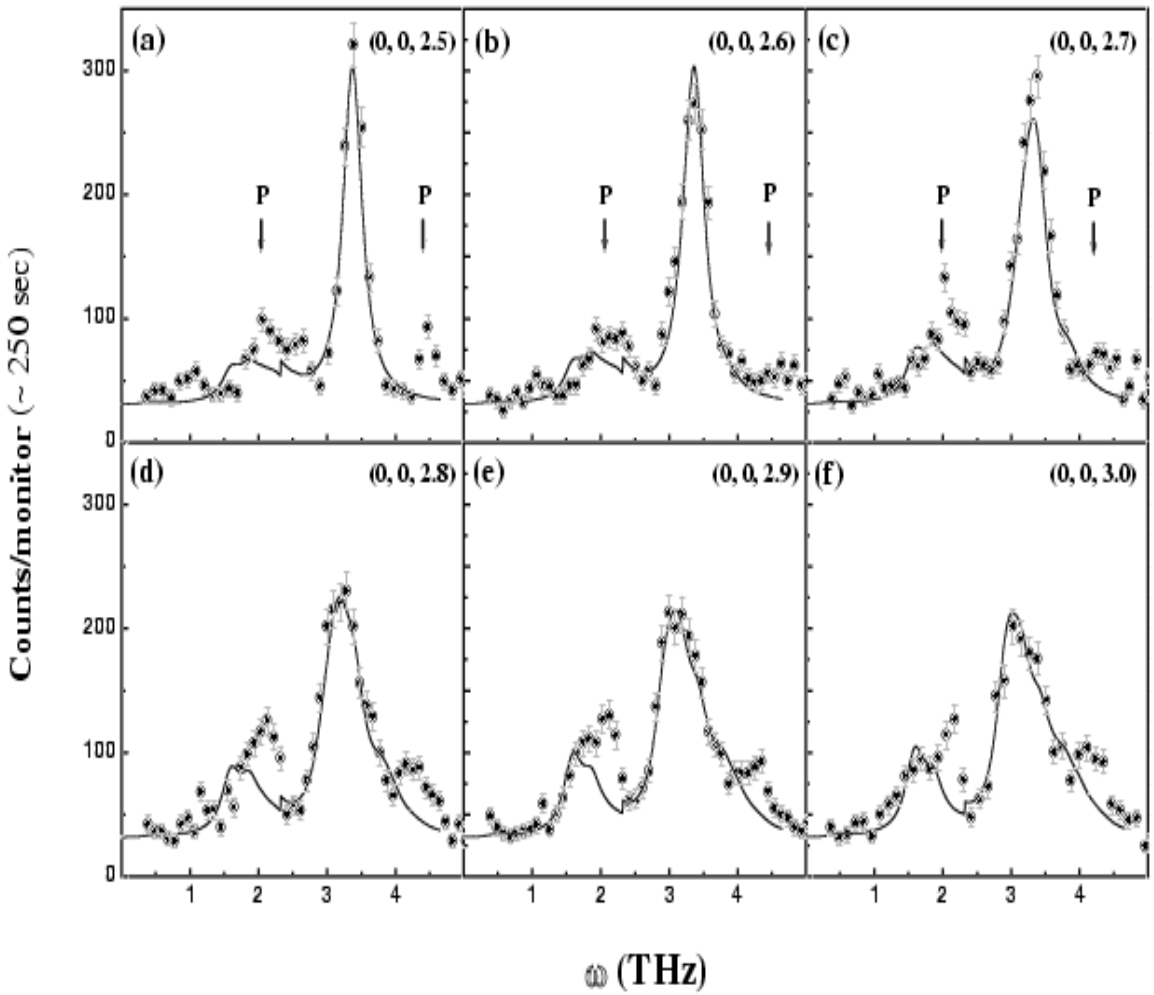

FIG. 1. Inelastic neutron scattering at several wave vectors of the form (0, 0, L), at 13.3 K, from $CsCo_{0.83}Mg_{0.17}Br_3$ are shown. The solid curves are a high quality fit to the data due to theoretical calculations of $S^{xx}(Q,\omega)$ given by Eq. (3). The parameters used in the calculations are J=1.55 THz, h=0.05J, ϵ=0.18J, Δ=0.1J, T=13.3 K.

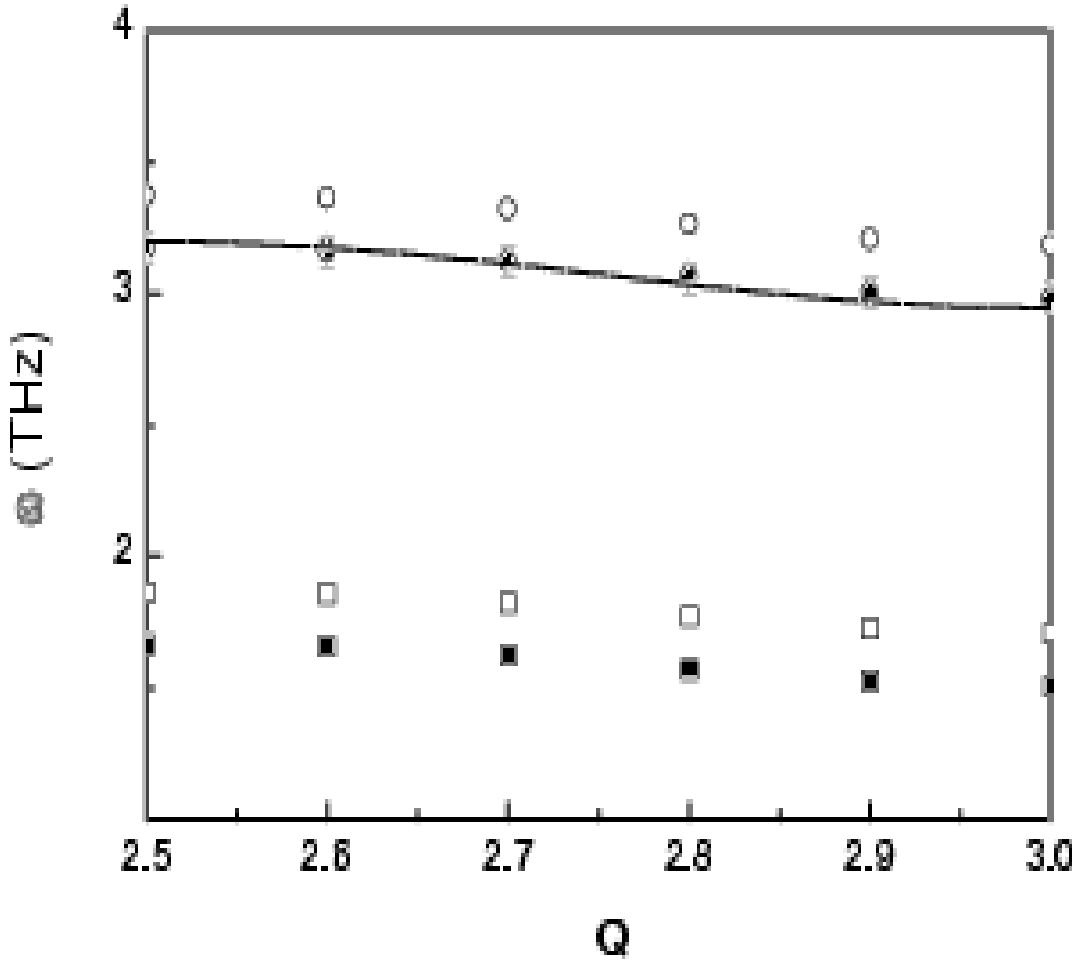

FIG. 2. The dispersion relation obtained from Fig. 1 for the $2J$ (J) mode is shown as the open circles (squares). The filled circles are these energies with the value $0.13J$ (0.2 THz) subtracted from open ones. The line corresponds to the IS theoretical expression, Eq. (2).

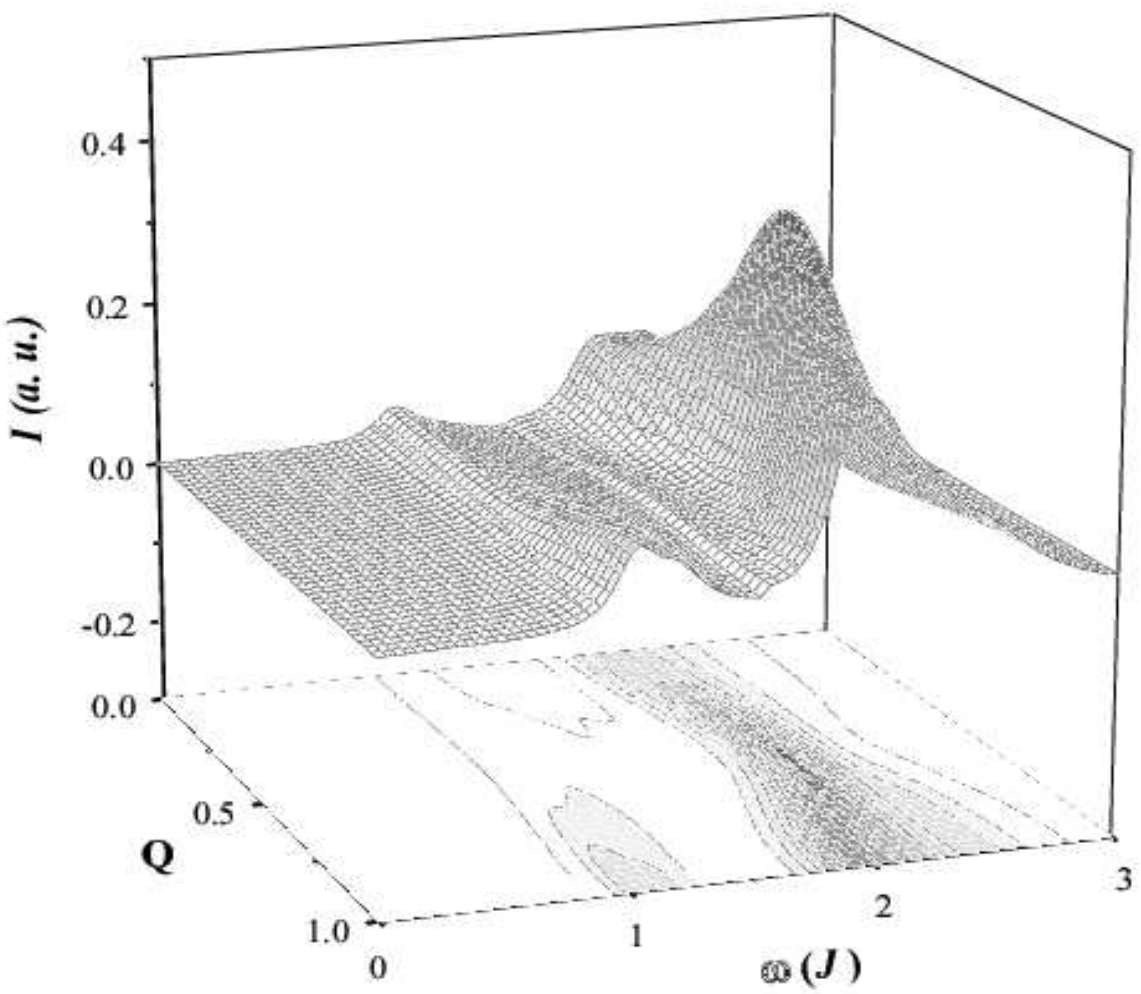

FIG. 3. The calculated two dimensional contour map of the spin wave spectra $S^{xx}(Q,\omega)$. The parameters used in the calculation are J=1.55 THz, h=0.05J, ϵ=0.18J, T=13.3 K and Δ=0.1J.

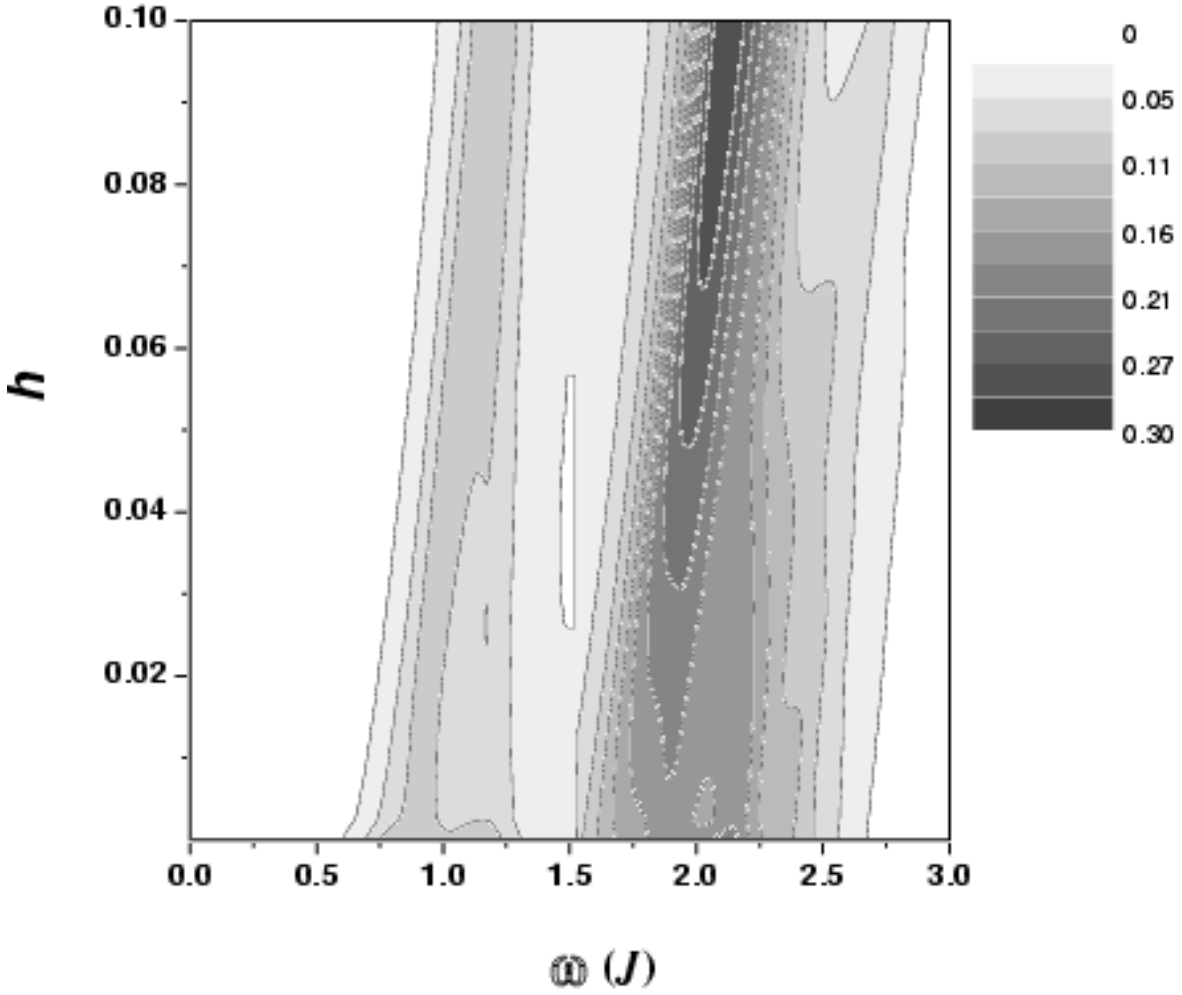

FIG. 4. The calculated two dimensional contour map of the spin wave spectra are shown as a function of ω and the staggered term h for the zone center wavevctor. The parameters in the calculation are those used in Fig. 3.